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We have studied processes in excited atoms driven by strong microwave radiation fields to gain a quantitative understanding of what are usually thought of as highly nonlinear processes. We have discovered that circularly polarized microwave fields produce ionization only at fields above 1/16n4, the much higher field required for static field ionization. We have also observed above threshold ionization at microwave frequencies and shown that in the microwave regime it is easily described by a classical theory, which ties together above threshold ionization and ponderomotive energy shifts. Finally, we have made significant progress toward observing collisions which begin and end under our control.

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by

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I. Introduction

Under the AFOSR sponsorship of this research program,"Structure and Dynamics of Excited States," we have been investigating atoms in strong radiation fields. Specifically, we have studied highly excited, or Rydberg, atoms in microwave fields which are easily made as strong as the coulomb field binding the Rydberg electron to the atom. This experimental approach has allowed us to study strong field, or equivalently high order multiphoton, processes in a much more quantitative fashion than can be done with high power pulsed lasers and ground state atoms. We have studyied both strong field processes involving one excited atom, multiphoton excitation and ionization, and collisions between two excited atoms in the presence of strong radiation fields. Our experiments have provided unambiguous data which often point clearly to useful ways of thinking about high field processes. In fact recent optical experiments with high powered lasers have shown that the point of view developed from our experiments is useful in interpreting their optical counterparts.[1,2]

II. Accomplishments during the period 1 November 1986 to 31 October 1987

During this period we made progress in studying the interactions of single atoms with microwave fields and high resolution collisional energy transfer.

Resonant Collisional Energy Transfer

During this period we have made several advances in the study of collisional energy transfer. The first of these was to finish the theoretical analysis of the radiative collisions in the strong field regime, in which many, up to four, microwave photons are absorbed in the collisions. A report of this work has been published in *Physical Review A* and is listed below as No. 1.

In preparation for more detailed experiments we chose to examine some of the systematic effects more carefully. One of these is the velocity dependence of the collisional resonance linewidth. This we have investigated using a velocity selected beam of K atoms. By restricting the velocity difference between atoms to a very small value we have been able to observe very narrow collisional resonances, 6 MHz wide. This resolution has in fact enable us to make a spectroscopic measurement using these collisions. Specifically we were able to determine the quantum defect of the K p states to an order of magnitude better than had been done previously. A report of this work has been published in *Physical Review Letters* and is listed below as No. 2.

Microwave Multiphoton Excitation and Ionization

During this period a report of the first measurements of microwave multiphoton resonances, in K atoms, was published in *Physical Review Letters* and is listed below as No. 3. This work led us to try to analyze it quantitatively, we were in fact able to show that these transitions can be thought os as multiphoton transitions produced by an oscillating field or as anticrossings of atomic states dressed by the microwave field.

We began the study of microwave ionization of Li, in an attempt to make the connection between our studies of alkalis and experiments with H. In the course of this work we discovered a very interesting effect, namely that the microwave ionization of Li is enormously enhanced by the addition of small, ~1V/cm static fields. This small static field in combination with a microwave field converts the Li Rydberg states into a quasi continuum enabling the ionization to proceed by a sequence of necessarily resonant steps.

Quasi Static Field Ionization

During the period of this grant a report of the field ionization of Na atoms down to $n = 8$ was published in *Physical Review A* and is listed below as No. 4. These measurements show in detail how field ionization actually occurs, and are the lowest lying states ever ionized in a controlled fashion using field ionization.

Publications

1. P. Pillet, R. Kachru, N. H. Tran, W. W. Smith and T. F. Gallagher, "Radiative Rydberg atom-Rydberg atom collisions in the strong field regime." Phys. Rev. A *36*, 1132 (1987).

2. R. C. Stoneman, M. D. Adams, and T. F. Gallagher, "Resonant Collision Spectroscopy of Rydberg Atoms", Phys. Rev. Lett. *58*, 1324 (1987).

3. L. A. Bloomfield, R. C. Stoneman and T. F. Gallagher, Microwave Multiphoton transitions

between Rydberg States of potassium," Phys. Rev. Lett. *57*, (1986).

4. J. L. Dexter and T. F. Gallagher, "Field ionization of the n=8-15 states of Sodium," Phys. Rev. A *35*, 2345 (1987).

5. T. F. Gallagher, "Microwave Excitation and Ionization", in *Photons and Continuum states of atoms and molecules*." N. K. Rahman, C. Gwidotti, and M. Allegrini eds. (Springer-Verlog, Heidelbrug, 1987).

III. Accomplishments during the period 1 November 1987 to 31 October 1988

We carried out experiments in both microwave ionization and excitation and collisional energy transfer. These projects are described in more detail below.

Collisional Energy Transfer

During this period we completed an interesting series of measurements on the dependence on the relative orientation of the relative collision velocity $\bar{v}$ and the tuning electric field $\bar{E}$ in the resonant collision process

$$\text{(1)} \qquad \text{Na ns} + \text{Na ns} \rightarrow \text{Na np} + \text{Na(n-1)p} \quad .$$

which is tuned to resonance by adjusting $|\bar{E}|$. While we find that the cross sections, integrated over the resonance width, are virtually the same for $\bar{v} \parallel \bar{E}$ and $\bar{v} \perp \bar{E}$, there is a distinct difference in the lineshapes. A collisional resonance corresponding to Eq. (1) with $\bar{v} \perp E$ has a Lorentzian lineshape, while the corresponding $\bar{v} \parallel \bar{E}$ resonance lineshape has a sharp drop at the center of the resonance. The origin of the lineshape can be understood by working out the theory for this collisional process. In theory the cross section actually vanishes at exact resonance, because the dipole-dipole interaction changes sign in the collision and the contributions to the transition amplitude from earlier and later parts of the collision exactly cancel. Off resonance the cancellation is not complete, and the cross section does not vanish. The resonance lineshape is analogous to that observed in a separated oscillatory field experiment with a 180^0 phase shift between the two fields. To our knowledge this is the first observation of intracollisional interference.

One of our interests in the study of resonant collisions such as those described above is that we can easily study collisions in which microwave photons are absorbed. We have in fact made a study of the n dependence of the collision process.

$$\text{Na ns} + \text{Na ns} + mh\nu \rightarrow \text{Na np} + \text{Na(m-1)p} + (m+1)h\nu \qquad (2)$$

which is again tuned into resonance with a static electric field. In the low microwave power regime, in which the process described by Eq. (2) is at least three times weaker than that described by Eq. (1) we find that the microwave field required scales as n^{-2}. We have compared this experimental result to values calculated using our earlier theoretical description of the process, and we find agreement between the measured and calculated fields to within 30 %.

Microwave Multiphoton Excitation and Ionization

We have completed an extensive set of resonant multiphoton excitation measurements and calculations which show for the first time the explicit connection between the time varying field[5] and multiphoton[6] points of view. A report of this work has been published in *Physical Review A* and is listed below as No. 1.

We have devoted a substantial effort the study of hydrogen-like microwave ionization. We began by studying Li, which has very small quantum defects and, correspondingly, small avoided crossings between Stark states of different n. As a result of the small avoided corssings Li atoms do not readily exhibit the $E = 1/3n^5$ Landau Zener form of ionization, which is common in Na and K. The small sizes of the avoided crossings ensures that the transitions are always off resonant. However, as we have found, a small (~ 1 V/cm) field converts the Stark states to a quasi continuum, ensuring that the transitions are resonant and allowing the $E = 1/3n^5$. A report of this work has been published in *Physical Review Letters* and is listed below as No. 2.

When Li atoms are not in a static field they ionize at a microwave field $E = 1/9n^4$, for n ~ 30, as do the hydrogenic Na $|m| = 2$ states and hydrogen itself. For n ~ 50 though the ionization occurs at a significantly lower field, $E \sim 3/n^5$ which corresponds to the field required to saturate the $n \rightarrow n+1$ transition between two extreme Stark states. We have studied this process experimentally in both Li and the Na $|m| = 2$ states and developed a very simple model to explain the results.

Finally, we began to study how microwaves ionize atoms in more detail. We began by asking whether or not the electron continues to absorb radiation above the ionization limit. In other words, does above threshold ionization occur with microwaves. By analyzing the energy dependence of the ejected electrons we have found that the electrons absorb enormous amounts of energy. Our observations are fit by a simple classial model which shows very clearly the connection between above threshold ionization and ponderomotive shift of the ionization limit. This connection is not particularly evident in previous theoretical work.

A summary of our work on microwave ioniztion and excitation was reported at the Fourth International Conference on Resonance Ionization at Gaithersburg in April, 1988, and at the Symposium on Atoms in Strong Fields at Grainau, West Germany in September, 1988. The contribution to the RIS conference is listed below as No. 3.

Publications:

1. R. C. Stoneman, D. S. Thomson, and T. F. Gallagher, "Microwave multiphoton transitions between Rydberg states of Potassium", Phys. Rev. A *37*, 1527 (1988).

2. P. Pillet, C. R. Mahon, and T. F. Gallagher, "Enhancement of microwave ionizaton by Quasicontinuum production", Phys. Rev. Lett. *60*, 21 (1988).

3. T. F. Gallagher, "Microwave Multiphoton excitation and ionization" in *Resonance Ionization Spectroscopy*, eds. T. B. Lucatorto and J. E. Parks (Institute of Physics, Bristol 1988).

IV. Accomplishments during the Period 1 November 1988 to 31 October 1989

During this period we focused our efforts on three areas: microwave ionization, above threshold ionization, and resonant collisional energy transfer. In the rest of this section we describe this work in more detail.

Microwave Ionization

In the past year we finished a study of the microwave ionization of both Na and Li atoms over the broad frequency range from 600 MHz to 30 GHz. A full report of this work is in preparation at this time, but a report of one aspect has been published in *Physical Review A* and is listed below as No. 1. This aspect is the comparison of the the microwave ionization of hydrogenlike Na and Li m=2 states to H itself. One point of view is that the microwave ionization of H can only be understood using the methods of classical chaos.[3] In Figure 1 we show the microwave ionization fields for the Li and Na m=2 states as well as the H states for principal quantum number n from 32 to 70. As shown in Figure 1 the data all lie together, especially if allowance is made for the fact that they are from three different experiments. What is more interesting, at least to us, is that these data can be understood in a simple fashion. At n=32 the data fall near the upper solid line, $E=\frac{1}{9n^4}$, which corresponds to the direct field ionization of the n state initially populated. As n increases, the levels become closer together, and it becomes increasingly possible for the microwave field to drive the single photon n to n+1 transition. It is a straightforward matter to show that the field required is given by $E=\frac{3}{n^5}$, which is the lower of the two solid lines in Figure 1. Clearly ionization will occur by whichever process requires the lower field. For the frequency we used, 15GHz, this model should break down at n=70. However, below this n the prediction of an $E=\frac{3}{n^5}$ threshold law is in exellent agreement with the data down to n=27, where the $E=\frac{1}{16n^4}$ law holds. In addition to the paper published on this subject, it was also the subject of an invited paper at the American Physical Society spring meeting.

The question of how atoms respond to intense polychromatic fields is one of some interest and importance. In fact a study of the microwave ionization of H by two frequencies was said to require a more sophisticated theory of classical chaos to explain the results.[4] We have studied the ionization of both hydrogenic and non hydrogenic states of Li and Na atoms by 14 and 15 GHz fields applied simulataneously. We found that in all cases what is crucial is the peak amplitude of the field, irrespective of whether it comes from one frequency or a combination of both. From our point of view this result is hardly a surprise, as it emerges easily from several rather different analyses of the problem. It appears that more sophisticated theories of chaos are not required to explain multifrequency ionization. A report of this work has been submitted to *Physical Review A*.

During this past year we finished the preparation of a manuscript on the microwave ionization of Ba. Our original interest was to determine if Ba, a two valence electron atom, exhibited the same sort of behaviour as the single valence electron alkali atoms. In fact, Ba exhibits precisely the same phenomena observed in the alkali atoms.[5,6] However the data are, at first glance, much more confusing since the Ba Rydberg states have many doubly excited valence state perturbers, which alter the quantum defects of the Ba Rydberg series, and hence the response of the Ba atoms to the microwave field. A report of this work has been published in *Zeitschrift fur Physik* and is listed below as No. 2.

Until the present all work on microwave ionization has been focused on the measurement of

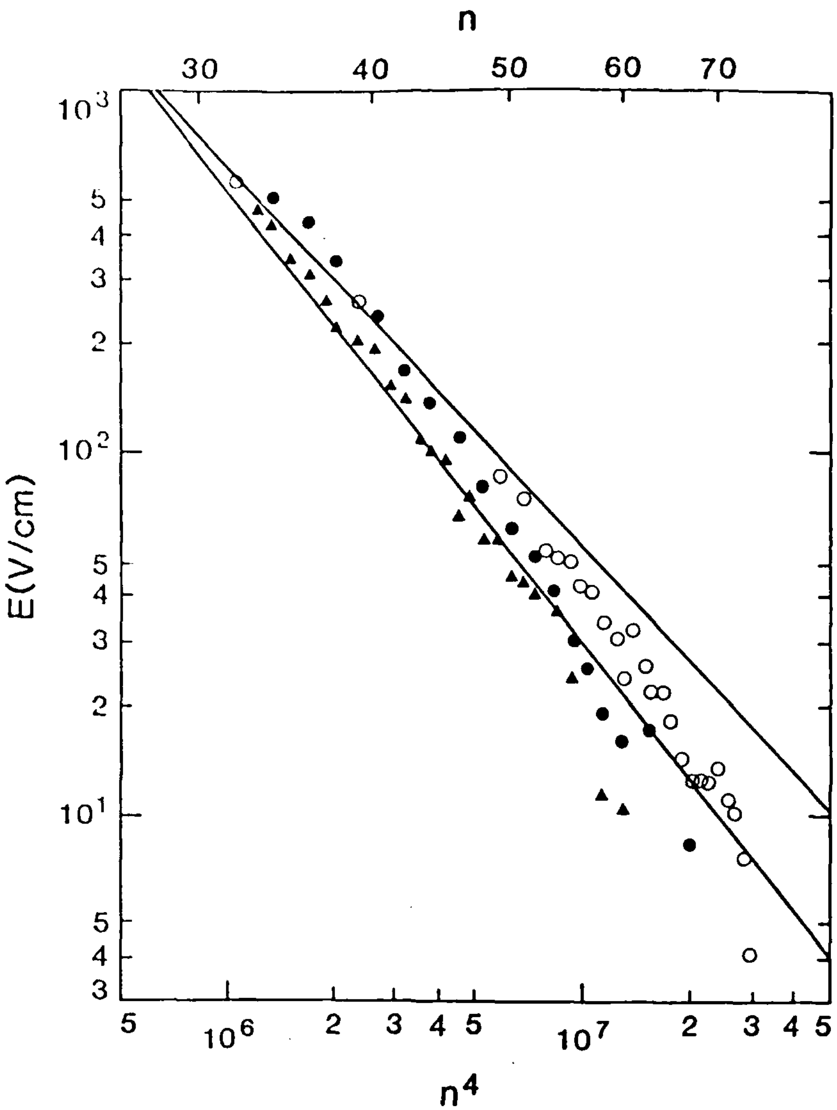

Figure 1. Microwave ionization threshold fields of Li (▲) and Na (●) both with m=2, and H (○). Also shown are the lines $E=1/9n^4$ (upper line) and $E=3/n^5$ (lower line), corresponding to the fields for direct and multistep ionization.

the threshold fields for ionizaton, in spite of the fact that it is not an instantaneous process but rather one which has a rate which depends on the microwave field. Precisely how fast microwave ionization occurs is still not known. To shed some light on this problem we have begun a detailed study of the rates for microwave ionization by measuring directly the time resolved electron signals from the microwave ionization. We have been able to measure rates as fast as 10^8 s^{-1}, which is near the limit of the time resolution imposed by the laser, and we have found that at low n the rates increase more rapidly with microwave power than at high n. This observation is not a complete surprise, for we had previously noticed that the high n ionization thresholds were not as sharp as the low n ones.

What did come as a surprise, however, was the fact that the rates for ionizing different states of the same n are very different. This phenomenon is shown in Figure 2, which shows that the states in the center of the Stark manifold ionize as much as a factor of ten more slowly than the states at the edge of the manifold. We explain this diiference in the following way. To ionize, the atom must make the n to n+1 transition, which occurs from the extreme blue Stark manifold state. An atom initially in a central Stark state can reach the extreme state by undergoing a transition at the zero field level crossing of all the ℓ>2 states. This crossing is traversed very rapidly, with the effect that the states are projected diabatically from the positive field states to the negative field states. The net effect is that the extreme upper states are projected mainly onto the extreme lower states and vice versa. The central states are projected mainly onto themselves. In other words the transitions from the central states to the extreme states occur slowly. In fact, the transitions from the central Stark states to the extreme states are slower than the sequence of n changing transitions that results in ionization. We are preparing a report of this aspect of the work for publication.

As a complementary study to the direct measurement of the microwave ionization rates we have been examining the effect of the shape and duration of the microwave pulse on both the probability of microwave ionization and the final state distribution subsequent to the exposure to the microwaves. These experiments are interesting in their own right and will test theoretical results on multiphoton ionization. One theory suggests that all the ionization is due to the changes in the field amplitude, not just to its presence.[7] Futhermore, numerical calculations of multiphoton processes are frequently carried out with a small number, roughly fifty, oscillations of the field.[8] Our experiments will test these theories. Preliminary results indicate that the most sensitive parameter is the pulse length. As the pulse length becomes shorter, the ionization probability drops sharply, in contradiction to the theory based on the changes in the field amplitude.

In optical multiphoton experiments there is observed to be a large difference in efficiency between linearly and circularly polarized light. Linear polarization is much more effective in driving miultiphoton processes, and the difference increases with the order of the process. In this past year we have begun the study of microwave ionization by circularly polarized fields. The approach is to set up two orthogonal linear polarized standing waves $\frac{\pi}{2}$ out of phase in a Fabry Perot cavity as shown in Figure 3. We have found a striking difference between linear and circular polarization. With linear polarization a field $E=\frac{1}{3n^5}$ is required, but with circular polarization a field $E=\frac{1}{16n^4}$ is required, a much stronger field which is of the same value as the static field required for ionization. The linear and circular polarization threshold dependences are shown in Figure 4.

The circular polarization results can be understood in a straightforward way. If the microwave field is in the x-y plane and rotates at frequency $\frac{\omega}{2\pi}$ about the z axis, we transform the

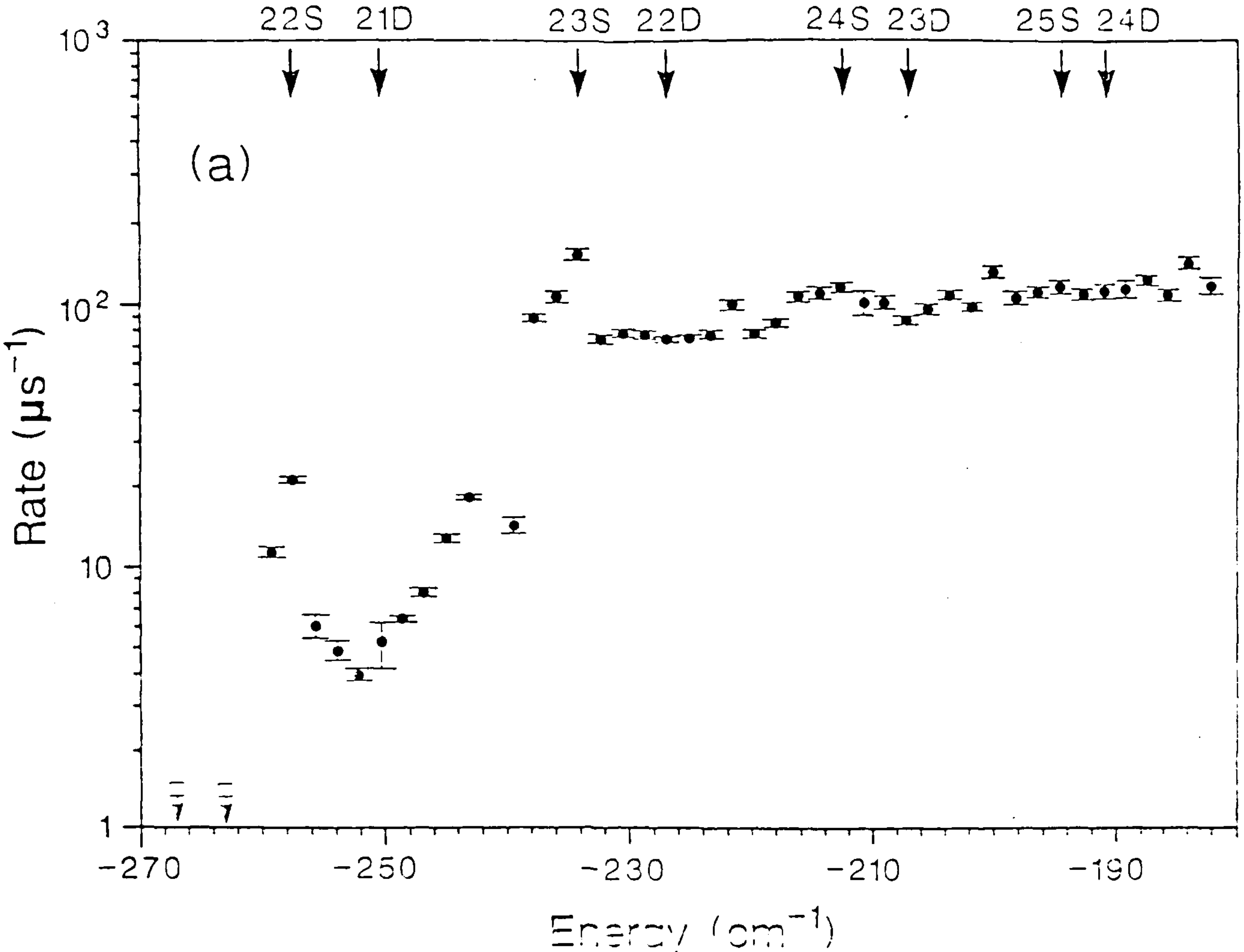

Figure 2. Microwave ionization rate vs binding energy as the exciting laser is tuned across several Stark manifolds. The 8 GHz microwave field amplitude is 310 V/cm. Note the much lower ionization rate at the location of the zero field 21d state, which corresponds to the center of the n=21 Stark manifold.

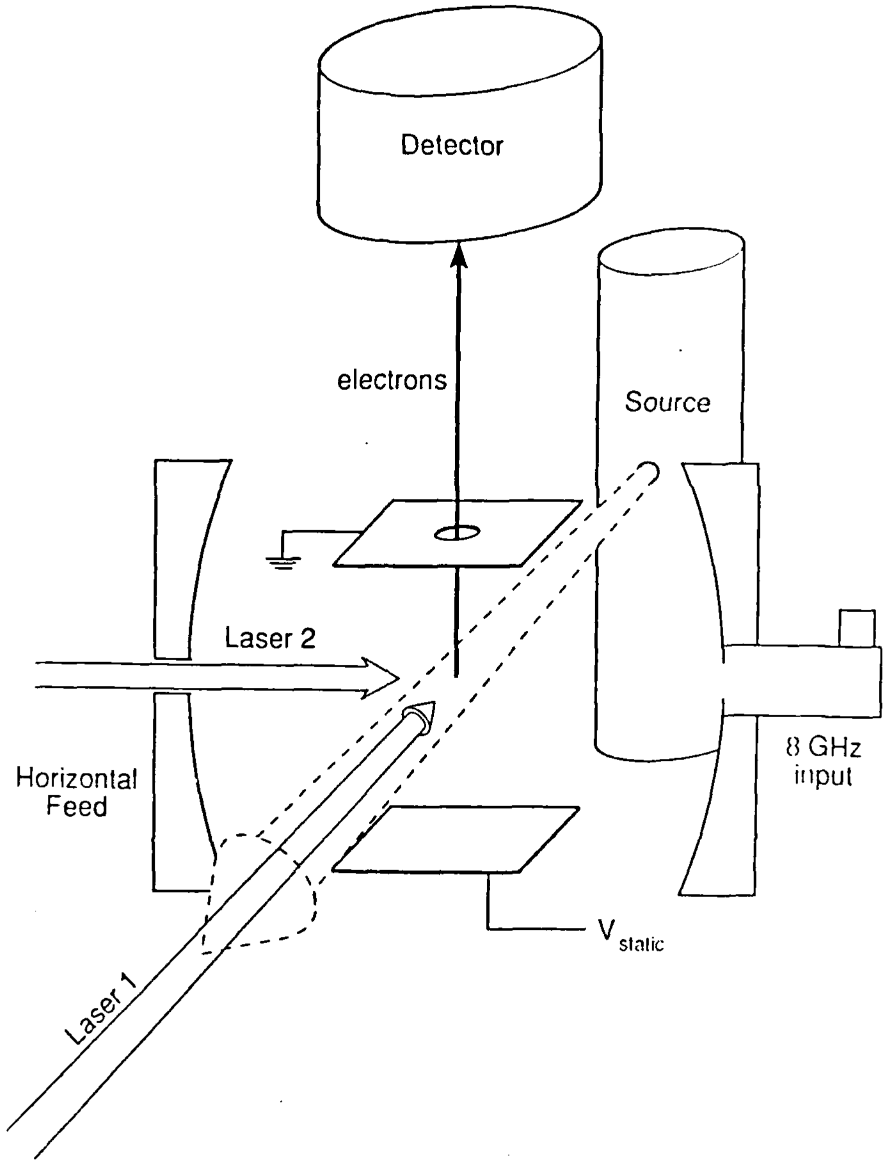

Figure 3. The apparatus used to measure microwave ionization in a circularly polarized field.

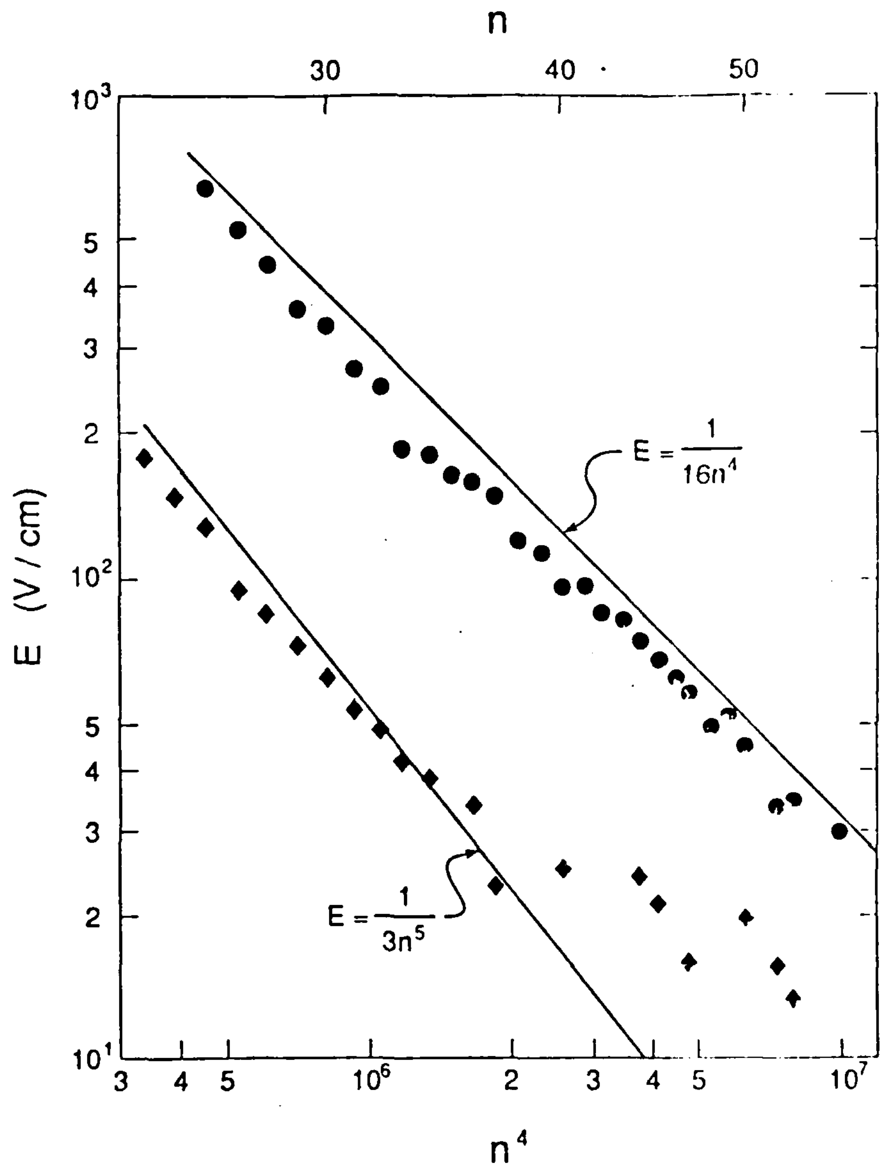

Figure 4. Ionization threshold fields for linear (♦) and circular (●) polarization as a function of n when the atoms are excited in the microwave field.

problem to a frame rotating with the microwave field.[9] This transformation does two things. The microwave field becomes a static field, and states of angular momentum m along the z axis are shifted in energy by $-m\hbar\omega$. In the rotating frame ionization occurs just as it does in a static electric field, at the classical ionization limit, $E=\frac{1}{16n^4}$. Although it is tempting to conclude that the ionization is exactly the same as static field ionization, it is not. In a circularly polarized field states of even or odd ℓ+m are coupled, while in a linearly polarized field any states of the same m are coupled. A report of this work has been submitted for publication to *Physical Review Letters* and is included here as Appendix E.

Above Threshold Ionization

In the optical regime the phenomenon of above threshold ionization(ATI), the process in which an atom absorbs more than the minimum number of photons required to effect multiphoton ionization, has been studied extensively. Furthermore it is empirically observed to be related to the ponderomotive shift of the ionization limit, the energy shift corresponding to the fact that when the electron is freed into an oscillating field it has an energy associated with the oscillation. This energy, $W=\frac{e^2E^2}{4m\omega^2}$, is the ponderomotive energy. The scaling of the ponderomotive energy as $\frac{1}{\omega^2}$ suggests that it should be very large at microwave frequencies. Accordingly, we have begun the study of ATI in the microwave regime by energy analyzing the energies of the electrons ejected by microwave ionization. We have found that the electrons are always observed to have energies between one and three times the ponderomotive energy. We have in addition developed a simple classical model which agrees with our data and has the great virtue of tying the ponderomotive shift to ATI in an obvious way. This work is described in papers published in *Physical Review Letters* and *Physical Review A*. These papers are listed below as Nos. 3 and 4.

The classical model predicts that the energy of the ejected electron is entirely dependent on the phase at which the electron becomes free of the atom. We are now in the process of testing this notion using a picosecond laser to create Rydberg atoms at well defined phases of the microwave field.

Resonant Collisional Energy Transfer

In the past year we published an account of our work on the resonant dipole-dipole collision process

$$\text{Na ns} + \text{Na ns} \rightarrow \text{Na np} + \text{Na n+1p}$$

which is tuned into resonance with an electric field. The field direction and the relative velocity of the two atoms are the two vectors that most naturally define the geometry of the collision. When these two vectors are aligned, all collisions are the same but for the impact parameter. In this case we observe a unique intracollisional interference, which is analogous to the interference pattern observed in the Ramsey method of separated oscillatory fields.[10] In our case the interference is due to the dipole-dipole interaction's changing sign twice during the collision. Since the dipoles are actually oscillating, the sign change of the dipole-dipole interaction is equivalent to a phase shift of π between the two radio frequency fields in a Ramsey separated oscillatory field experiment. In either case the transition probability vanishes at reesonance, but not away from resonance. An example of a collisional resonance showing the interference is shown in Figure 5. A paper describing this work has been published in *Physical Review A*. It is listed below as No. 5

The resonant dipole-dipole collisions provide a good opportunity to study collisions in which photons are absorbed during the collisions. In a previous work we presented a theory to describe

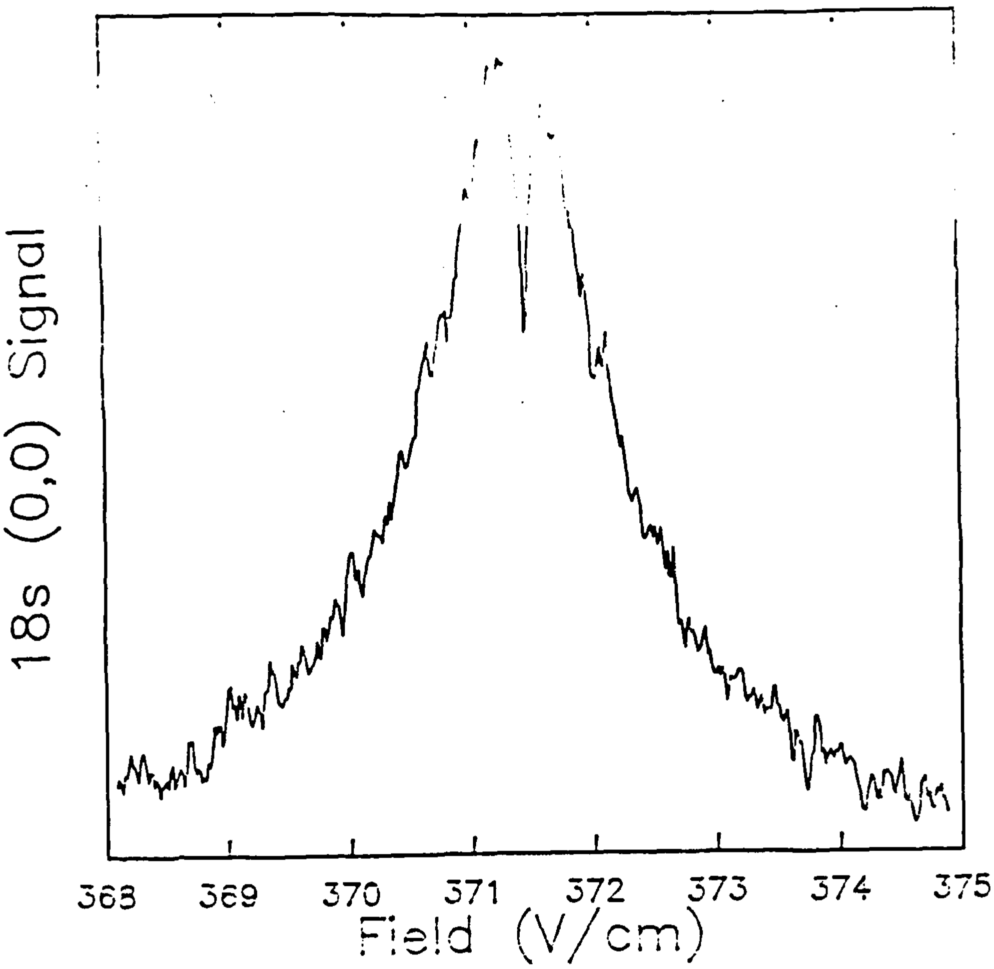

Figure 5. Experimentally observed collisional resonance for the process Na 18s +Na 18s- Na 18p +Na 17p when the collision velocity and the static field are aligned. The interference dip in the center of the resonance is apparent.

such radiatively assisted collisions.[11] We have now completed a study of the pricipal quantum number scaling of the microwave power required to induce collisions in which one photon is absorbed during the collision. As the principal quantum number n is increased the dipole moments increase, and less microwave power should be required. To our knowledge this experiment is the first such systematic test of any theory of radiatively assisted collisions, and the results of the experiment are in good, 20, agreement with the theory. A report of this work has been published in *Physical Review A*, and it is listed below as No. 6.

We have velocity selected a beam of K atoms in an attempt to observe collisions with linewidths as narrow as the inverse of the observation time. These collisions are transform limited in the sense that it is impossible for the collisional resonances to be any narrower. Another interesting aspect of such collisions is that we know when the begin and end. To our knowledge this has never been possible before with an atomic collision. Knowing when individual collisions begin and end we can perturb the collisions at well defined times during the collision and observe the effect on the collisional resonance lineshape.

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1. T. F. Gallagher, C. R. Mahon, P. Pillet, P. Fu, and J. B. Newman, "Microwave Ionization of Hydrogen like Li and Na atoms," Phys. Rev. A *39*, 4545 (1989).

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V. Conclusion

With the support of this grant we have made substantial progress in our understanding of several aspects of microwave ionization and resonant collisional energy transfer. We have shown the connection between multiphoton transitions and transitions driven by a time varying quasi static field, and we have shown how excitation through a quasi continuum depends critically upon the distributions of the energy levels. Finally we have observed collisions between excited atoms of longer duration than ever before observed, and we have used the control over these collisions to investigate atomic collisions in more detail than previously observed.

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